

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

ARIGA Toshio, et al.

Serial No.: 09/926,578

Filed: November 20, 2001



Group Art Unit: 1713

Examiner: Patrick Dennis Niland

For: AGENT IMPARTING IMPACT RESISTANCE AND POLYESTER  
COMPOSITION CONTAINING THE AGENT

DECLARATION UNDER 37 CFR §1.132

Honorable Commissioner  
of Patents and Trademarks  
Washington, D.C. 20231

Sir:

I, Shoji Imamura of 2-15-13, Osakidai, Sakura, Chiba, Japan,  
do declare as follows:

I finished my bachelor course at the Tokyo University of Science in 1990 and master course at the Tokyo Institute of Technology in 1992, respectively, and I was awarded the degrees of B.A. and M.A. Since April 1, 1992, I have been employed by Dainippon Ink and Chemicals, Inc., the assignee of the above-identified application. Since April 1, 1993, I have been engaged in CP-6 Project (especially lactic acid-based copolymerization) at Central Research Laboratories of Dainippon Ink and Chemicals, Inc.

In order to demonstrate that the composition according to the present invention exhibits unexpected results, I performed the following experiments:

EXPERIMENTS

## 1. Purpose of experiments

The Examiner asserts that lactic acid polyester (III) of the present invention is the same as a "lactic acid-based polyester" disclosed in the citations Imamura et al., Kakizawa et al., Ebato et al., and Kolstad et al., and further asserts that the polyester composition according to the present invention, which essentially contains a polylactic acid (V) and an agent imparting impact resistance (IV) which is composed of a lactic acid polyester (I II), is the same as the "lactic acid-based polyester". However, these assertions are not correct.

In order to clearly show differences between lactic acid polyester (III) and the polyester composition according to the present invention, and the "lactic acid-based polyester", and to clarify the situation, the following experiments were carried out. The experiments clearly show that it is difficult to form a film by using a lactic acid-based polyester alone, and therefore a molded article composed of the lactic acid-based polyester alone is difficult to obtain.

The following experiments show that, even if a lactic acid-based polyester used in the citations, which can form a film, is selected and is used alone, when the amount of polyester unit (II) in the lactic acid-based polyester is equal to the amount of polyester unit in the polyester composition according to the present invention, the polyester composition according to the present invention has superior properties to the lactic acid-based polyester. That is, the following experiments show that, even if the content of polyester (II) which yields superior properties to the polyester composition is determined to be less than the amount of polyester units in the lactic acid-based polyester, the polyester composition of the present invention exhibits excellent properties which are equal to or superior to the properties exhibited by a lactic acid-based polyester alone.

## 2. Experiments

Aliphatic polyester (A-1) was prepared according to Reference Example 1 (Synthesis of Aliphatic Polyester A-1) on

page 24 of the present application. That is, 1 mol equivalent of "Empol 1061" and 1.4 mol equivalents of propylene glycol were charge into a 50 L reaction tank equipped with a stirrer, rectifying tube, and gas feed tube followed by heating and stirring while raising the temperature at the rate of 10°C per hour starting at 150°C under a nitrogen flow. The temperature was raised to 220°C while distilling off the formed water, and after 2 hours, transesterification catalyst, titanium tetrakisopropoxide was added at 70 ppm and the pressure was lowered to 0.1 KPa followed by stirring for 3 hours to obtain aliphatic polyester (A-1) having number average molecular weight ( $M_n$ ) as determined by polystyrene conversion using GPC of 18,000, and a weight average molecular weight ( $M_w$ ) of 30,000.

An agent imparting impact resistance (or a lactic acid polyester) (C-1) was prepared according to Production Example 1 (Synthesis of Agent Imparting Impact Resistance) on page 28 of the present application. That is, 50 parts by weight of aliphatic polyester (A-1) and 50 parts by weight of L-lactide were placed in a separable flask and melted at 180°C. After the melted liquid became uniform, tin octanoate was added at 200 ppm followed by stirring for 3.5 hours at 180°C. Following completion of polymerization, ethylhexanoic phosphate was added at 500 ppm to obtain lactic acid polyester having a number average molecular weight ( $M_n$ ) as determined by polystyrene conversion using GPC of 25,000, and a weight average molecular weight ( $M_w$ ) of 50,000. This preparation is Example 2.

A polymer blend (that is, a polyester composition) (P-1) was prepared according to Example 1 (Production of Polymer Blend P-1) on page 33 of the present application. That is, 85 parts by weight of "Lact y" and 15 parts by weight of agent imparting impact resistance (C-1) were kneaded for 10 minutes while heating at 190°C using a Laboplast Mill Mixer to obtain a polymer blend (P-1). This preparation is Example 1.

Production Example 1' - Synthesis of Lactic Acid Polyester (P-1')

Materials and the production method used in Production Example 1 to prepare lactic acid polyester (C-1) were adopted, except that 7.5 parts by weight of aliphatic polyester (A-1) in polymer blend (P-1) were used.

7.5 parts by weight of aliphatic polyester (A-1) in polymer blend (P-1) and 92.5 parts by weight of L-lactide were charged into a separable flask, and melted at 180°C. After the melted liquid became uniform, tin octanoate was added at 200 ppm followed by stirring for 3.5 hours at 180°C. Following completion of polymerization, ethylhexanoic phosphate was added at 500 ppm to obtain lactic acid polyester (P-1') having a weight average molecular weight ( $M_w$ ) of 130,000, and a number average molecular weight ( $M_n$ ) of 68,000. This preparation to Example 3.

Lactic acid polyester (C-1), polymer blend (P-1), and lactic acid polymer (P-1') prepared by the above Examples 1 to 3 were evaluated by Test Example 1 (Evaluation of Polymer Blends) and Test Example 2 (Dupont Impact Strength, Haze value, and Number of Days Until Start of Bleedout of Film) on page 37 of the present application.

Results and evaluations are shown in Table 1.

Example 1

Polymer blend (P-1) obtained by kneading polylactic acid (PLA) and lactic acid polyester (C-1) has formed a film having superior properties.

Example 2

Lactic acid polyester (C-1) was alone used to mold an Izod test piece by injection molding; however, the molded article was not easily separated from the die, the surfaces of the obtained molded article were rough, and the obtained molded article was distorted. As a result, the Izod test could not be carried out. Furthermore, lactic acid polyester (C-1) was alone used to try to form a film; however, the surface thereof has very sticky, and therefore, a film could not be formed.

Example 3

Though a film was produced using lactic acid polyester (P-1') alone, properties thereof were inferior to lactic acid polyester (P-1).

### 3. Results

Lactic acid polyester (C-1) obtained by Example 2 could not form a film and could not form a molded article. Furthermore, an Izod test piece was prepared by injection molding using lactic acid polyester (C-1) alone; however, the test piece was very flexible, so that it is difficult to use for injection molding of thin articles.

In Example 3, lactic acid polyester (P-1') was prepared using polyester unit (II) which has the same amount of polyester unit (II) contained in lactic acid polyester (P-1). However, Izod impact strength of lactic acid polyester (P-1') was one fifth that of lactic acid polyester (P-1), which is not strong. Furthermore, lactic acid polyester (P-1') shows remarkably inferior effects of decreasing storage modulus of elasticity, and shows double the haze value (%) for measurement of transparency.

In view of the above results, it is clear that the polyester composition according to the present invention prepared by essentially mixing a polylactic acid with an agent imparting impact resistance composed of a lactic acid polyester has properties which are superior to those of conventional lactic acid-based polyester.

Table 1

		Ex. 1	Ex. 2	Ex. 3
Polymer	Polyester used	PLA	C-1	P-1'
	Molecular weight (Mw/Mn) ( $\times 10,000$ )	25/16	5/2.5	13/6.8
	Charged amount (parts by weight)	85	-	-
Agent imparting impact resistance	Polymer name	C-1	-	-
	Charged amount (parts by weight)	15	-	-
Polymer blend	Blend name	P-1	C-1	P-1'
	Tg ( $^{\circ}\text{C}$ )	57	51	53
	Mp ( $^{\circ}\text{C}$ )	170	162	167
	Storage modulus of elasticity (GPa) 20 $^{\circ}\text{C}$	1.5	Molded article not formed	2.8
	Izod impact strength (KJ/m $^2$ )	15	-	3
250 $\mu\text{m}$ film	Haze (%)	10	Film not formed	20
	Dupont impact strength (J)	0.31	-	0.18
	Number of days until start of bleed out	1 year or more	-	200 days

For Declaration, add:

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

March 16, 2004  
Date

Shouji Inamura  
(Name)